Supporting Information

Anisotropic shock sensitivity of cyclotrimethylene trinitramine (RDX) from compress and shear reactive dynamics

Qi An[1], Yi Liu[1,2*], Sergey V. Zybin[1], Hyungjun Kim[3], and William A. Goddard III[1*]

[1]*Materials and Process Simulation Center, 139-74, California Institute of Technology, Pasadena, California 91125, USA*
[2]*School of Materials Science and Engineering, University of Shanghai for Science and Technology, Shanghai 200093, P. R. China*
[3]*Graduate School of Energy Environment Water Sustainability (EEWS), Korea Advanced Institute of Science and Technology (KAIST), Daejeon, 305-701, Republic of Korea*

Table S1. Bond order cut-off values for different atom pairs. *BondFrag* program uses these values as a default parameter set (can be adjusted by the user) to determine molecular fragments.

	C	**H**	**O**	**N**
C	0.55	0.40	0.80	0.30
H		0.55	0.40	0.55
O			0.65	0.55
N				0.45

Table S-2. Slip systems considered based on minimized systems with 10% compression for (100) shock. Resolved shear stress (RSS), angle of shock plane/slip plane (φ) and angle of shock direction/slip direction (ψ) after minimization of 10% compression under (100) shock. Since no slip systems were observed for (100) shock, the two slip systems with large RSS and good angles (close to 45 degree) marked by asterix were selected for CS-RD simulations. Table 1 show that these two directions lead to very similar CS-RD results. The stress tensor (Pxx, Pyy, Pzz, Pxy, Pyz, Pxz) = (4.14, 1.52, 2.42, 0.01, -0.02, -0.01) GPa is used to calculate RSS.

Slip plane	Slip direction	φ	ψ	RSS (GPa)
*{110}	<1-10>	45.54	44.46	1.31
*{-110}	<110>	134.46	44.46	1.31
{110}	<111>	45.54	53.55	1.08
{210}	<1-20>	26.70	63.00	1.06
{120}	<-210>	63.86	153.86	1.04
{101}	<10-1>	47.00	43.00	0.86
{20-1}	<102>	28.20	61.80	0.71
{2-10}	<001>	26.70	90.00	0.01

Table S-3. Slip systems considered based on minimized systems with 10% compression for (210) shock. Resolved shear stress (RSS), angle of shock plane/slip plane (φ) and angle of shock direction/slip direction (ψ) after minimization of 10% compression under (210) shock. The six slip systems marked by asterix were selected for CS-RD simulations. These five cases with low RSS were selected because they had previously been suggested as possible slip systems for (210) shock. The CS-RD finds that the top selected slip system is preferred. The stress tensor (Pxx, Pyy, Pzz, Pxy, Pyz, Pxz) = (3.38, 1.72, 1.97, 0.45, 0.00, 0.00) GPa is used to calculate RSS.

Slip plane	Slip direction	φ	ψ	RSS (GPa)
*{120}	<-210>	33.82	123.82	0.94
*{010}	<100>	57.83	32.17	0.59
*{021}	<100>	67.71	32.17	0.53
*{0-21}	<100>	118.29	32.17	0.53
{102}	<20-1>	68.33	39.56	0.51
{100}	<010>	27.00	63.00	0.45
*{011}	<100>	68.16	32.17	0.41
*{0-11}	<100>	111.84	32.17	0.41
{100}	<011>	27.00	71.01	0.32
{100}	<012>	27.00	78.02	0.20
{021}	<0-12>	67.71	101.98	0.08

Table S-4. Slip systems considered based on minimized systems with 10% compression for (111) shock. Resolved shear stress (RSS), angle of shock plane/slip plane (φ) and angle of shock direction/slip direction (ψ) after minimization of 10% compression under (111) shock. The seven slip systems marked by asterix were selected for CS-RD simulations. These seven slip systems had previously been suggested as possible slip systems for (111) shock. The CS-RD finds that the second selected slip system is preferred. The stress tensor (Pxx, Pyy, Pzz, Pxy, Pyz, Pxz) = (2.04, 1.18, 2.13, 0.14, 0.39, 0.16) GPa is used to calculate RSS.

Slip plane	Slip direction	φ	ψ	RSS (GPa)
*{011}	<100>	28.70	61.30	0.38
{001}	<100>	48.76	61.30	0.38
{001}	<110>	48.76	41.80	0.36
{100}	<011>	55.94	34.19	0.36
*{021}	<100>	33.16	61.30	0.33
*{010}	<001>	51.94	54.63	0.19
*{010}	<100>	51.94	61.30	0.18
*{0-11}	<100>	100.71	118.70	0.16
*{001}	<010>	48.76	56.78	0.10
*{0-21}	<100>	104.87	118.70	0.02

Table S-5. Slip systems considered based on minimized systems with 10% compression for (110) shock. Resolved shear stress (RSS), angle of shock plane/slip plane (φ) and angle of shock direction/slip direction (ψ) after minimization of 10% compression under (110) shock. The three slip systems marked by asterix were selected for CS-RD simulations. These three slip systems had previously been suggested as possible slip systems at ambient conditions. The CS-RD finds that the top selected slip system is preferred. The stress tensor (Pxx, Pyy, Pzz, Pxy, Pyz, Pxz) = (2.52, 1.63, 1.89, 0.30, 0.00, -0.01) GPa is used to calculate RSS.

Slip plane	Slip direction	φ	ψ	RSS (GPa)
*{010}	<100>	38.48	51.52	0.39
*{021}	<100>	45.44	51.52	0.35
{0-21}	<100>	134.56	51.52	0.35
{100}	<010>	45.54	44.46	0.30
{101}	<10-1>	62.41	62.10	0.30
{010}	<101>	38.48	62.10	0.28
*{011}	<100>	56.19	51.52	0.27
{0-11}	<100>	123.81	51.52	0.27
{100}	<011>	45.54	59.78	0.21
{001}	<100>	90.00	51.52	0.00
{001}	<010>	90.00	44.46	0.00
{100}	<001>	45.54	90.00	0.00

Table S-6. Slip systems considered based on minimized systems with 10% compression for (120) shock. Resolved shear stress (RSS), angle of shock plane/slip plane (φ) and angle of shock direction/slip direction (ψ) after minimization of 10% compression under (120) shock. The four slip systems marked by asterix were selected for CS-RD simulations. Three cases were selected despite a low RSS because they had previously been suggested as possible slip systems in ambient conditions. The CS-RD finds that the second selected slip system is preferred. The stress tensor (Pxx, Pyy, Pzz, Pxy, Pyz, Pxz) = (1.48, 2.18, 1.92, 0.55, -0.03, -0.03) GPa is used to calculate RSS.

Slip plane	Slip direction	φ	ψ	RSS (GPa)
*{-120}	<210>	44.64	45.36	0.65
{100}	<010>	63.86	26.14	0.55
*{010}	<100>	21.68	68.32	0.49
{-110}	<110>	62.91	27.09	0.49
{201}	<010>	67.65	26.14	0.46
*{0-21}	<100>	146.87	68.32	0.45
{021}	<100>	33.13	68.32	0.43
{100}	<0-11>	63.86	128.57	0.40
*{0-11}	<100>	132.04	68.32	0.37
{011}	<100>	47.96	68.32	0.33
{012}	<100>	64.63	68.32	0.20
{102}	<-201>	80.03	70.14	0.19
{102}	<010>	80.03	26.14	0.19

Table S-7. Slip systems considered based on minimized systems with 20% compression for (100) shock. Resolved shear stress (RSS), angle of shock plane/slip plane (φ) and angle of shock direction/slip direction (ψ) after minimization of 20% compression under (100) shock. Since no slip systems were observed for (100) shock, four slip systems with large RSS and good angle (close to 45 degree) marked by asterix were selected for CS-RD simulations. The CS-RD finds that the third selected slip system is preferred. The stress tensor (Pxx, Pyy, Pzz, Pxy, Pyz, Pxz) = (10.48, 7.12, 5.31, 0.02, 0.03, -0.15) GPa is used to calculate RSS.

Slip plane	Slip direction	φ	ψ	RSS (GPa)
*{101}	<10-1>	43.63	46.37	2.58
*{-101}	<101>	136.37	46.37	2.58
*{110}	<1-10>	42.17	47.83	1.67
*{-110}	<110>	137.83	47.83	1.67
{120}	<-210>	61.10	151.10	1.45
{210}	<1-20>	24.37	65.64	1.19
{130}	<-310>	69.80	159.8	1.14
{310}	<1-30>	16.80	73.20	0.85
{2-10}	<001>	24.37	90.00	0.13

Table S-8. Slip systems considered based on minimized systems with 20% compression for (210) shock. Resolved shear stress (RSS), angle of shock plane/slip plane (φ) and angle of shock direction/slip direction (ψ) after minimization of 20% compression under (210) shock. The six slip systems marked by asterix were selected for CS-RD simulations. Five cases with low RSS were selected because they had previously been suggested as possible slip systems. The CS-RD finds that the top selected slip system is preferred. The stress tensor (Pxx, Pyy, Pzz, Pxy, Pyz, Pxz) = (8.70, 5.19, 5.57, -0.12, 0.09, 0.04) GPa is used to calculate RSS.

Slip plane	Slip direction	φ	ψ	RSS (GPa)
*{120}	<-210>	30.77	126.88	1.61
*{010}	<100>	54.72	35.29	0.54
*{021}	<100>	58.80	35.29	0.52
*{0-21}	<100>	121.20	35.29	0.45
*{011}	<100>	65.73	35.29	0.44
*{0-11}	<100>	114.27	35.29	0.33
{021}	<0-12>	58.80	100.68	0.20
{100}	<010>	24.37	65.64	0.12
{100}	<011>	24.37	72.99	0.02
{100}	<012>	24.37	79.32	0.02

Table S-9. Slip systems considered based on minimized systems with 20% compression for (111) shock. Resolved shear stress (RSS), angle of shock plane/slip plane (φ) and angle of shock direction/slip direction (ψ) after minimization of 20% compression under (111) shock. The six slip systems marked by asterix were selected for CS-RD simulations. These six slip system had previously been suggested as possible slip systems. {0-11}/<100> slip system is not selected due to the small RSS although it was suggested by a previous study. The CS-RD finds that the second selected slip system is preferred. The stress tensor (Pxx, Pyy, Pzz, Pxy, Pyz, Pxz) = (7.01, 5.85, 5.77, 0.83, 0.72, 0.45) GPa is used to calculate RSS.

Slip plane	Slip direction	φ	ψ	RSS (GPa)
*{011}	<100>	25.95	64.05	1.16
*{021}	<100>	30.15	64.05	1.12
{001}	<110>	45.40	45.17	1.10
{100}	<011>	52.75	37.38	1.10
{001}	<100>	45.40	64.05	0.89
*{010}	<100>	47.71	64.05	0.88
{100}	<001>	52.75	57.74	0.72
*{010}	<001>	47.71	57.74	0.55
*{001}	<010>	45.40	64.05	0.55
*{0-21}	<100>	106.62	115.95	0.40
{0-11}	<100>	86.87	115.95	0.05

Table S-10. Slip systems considered based on minimized systems with 20% compression for (110) shock. Resolved shear stress (RSS), angle of shock plane/slip plane (φ) and angle of shock direction/slip direction (ψ) after minimization of 20% compression under (110) shock. The four slip systems marked by asterix were selected for CS-RD simulations. Three of these cases were selected despite a low RSS because they had previously been suggested as possible slip systems at ambient conditions. The CS-RD finds that the second selected slip system is preferred. The stress tensor (Pxx, Pyy, Pzz, Pxy, Pyz, Pxz) = (9.93, 6.00, 4.53, 0.26, 0.02, 0.00) GPa is used to calculate RSS.

Slip plane	Slip direction	φ	ψ	RSS (GPa)
*{101}	<10-1>	59.55	64.80	2.63
*{010}	<100>	35.24	54.76	1.07
*{021}	<100>	42.07	54.76	0.98
{0-21}	<100>	137.93	54.76	0.97
*{011}	<100>	53.00	54.76	0.80
{010}	<101>	35.24	64.80	0.79
{0-11}	<100>	127.00	54.76	0.78
{100}	<010>	42.17	47.83	0.26
{100}	<011>	42.17	62.63	0.19
{010}	<001>	35.24	90.00	0.00
{001}	<100>	90.00	54.76	0.02
{001}	<010>	90.00	47.83	0.00
{100}	<001>	42.17	90.00	0.02

Table S-11. Slip systems considered based on minimized systems with 20% compression for (120) shock. Resolved shear stress (RSS), angle of shock plane/slip plane (φ) and angle of shock direction/slip direction (ψ) after minimization of 20% compression under (120) shock. The four slip systems marked by asterix were selected for CS-RD simulations. Three cases were selected despite a low RSS because they had previously been suggested as possible slip systems at ambient conditions. The CS-RD finds that the second selected slip system is preferred. The stress tensor (Pxx, Pyy, Pzz, Pxy, Pyz, Pxz) = (5.11, 5.99, 4.74, 1.26, 0.03, -0.05) GPa is used to calculate RSS.

Slip plane	Slip direction	φ	ψ	RSS (GPa)
*{-120}	<210>	41.28	48.73	1.33
{100}	<010>	61.10	28.90	1.26
{201}	<010>	22.66	28.90	1.07
*{010}	<100>	19.46	70.54	1.05
*{021}	<100>	30.12	70.54	0.98
{-110}	<110>	60.08	29.92	0.97
{0-21}	<100>	149.88	70.54	0.95
*{011}	<100>	44.59	70.54	0.82
{100}	<0-11>	61.102	125.33	0.81
{0-11}	<100>	135.41	70.54	0.77
{012}	<100>	61.92	70.54	0.56
{102}	<010>	78.81	28.90	0.46
{110}	<1-10>	14.26	104.26	0.36
{102}	<-201>	78.81	72.20	0.09

Table S-12. Temperatures (at 2 ps), NO_2/RDX ratio (at 4 ps), shear stress overshoot (τ_{max}- τ_c), and strain energy density W (integration of stress up to 2 ps) for 30% compression-shear simulations for three shock directions.

Sensitivity	Shock plane	Slip system	T (K)	NO_2/RDX (%)	$\tau_{max} - \tau_c$ (GPa)	W (GPa)
sensitive	(100)	*{-110}<110>	1186	18.8	4.31	2.25
insensitive	(111)	*{021}<100>	806	3.85	2.23	1.98
insensitive	(110)	*{010}<100>	650	1.13	1.02	1.33

Figure S-1

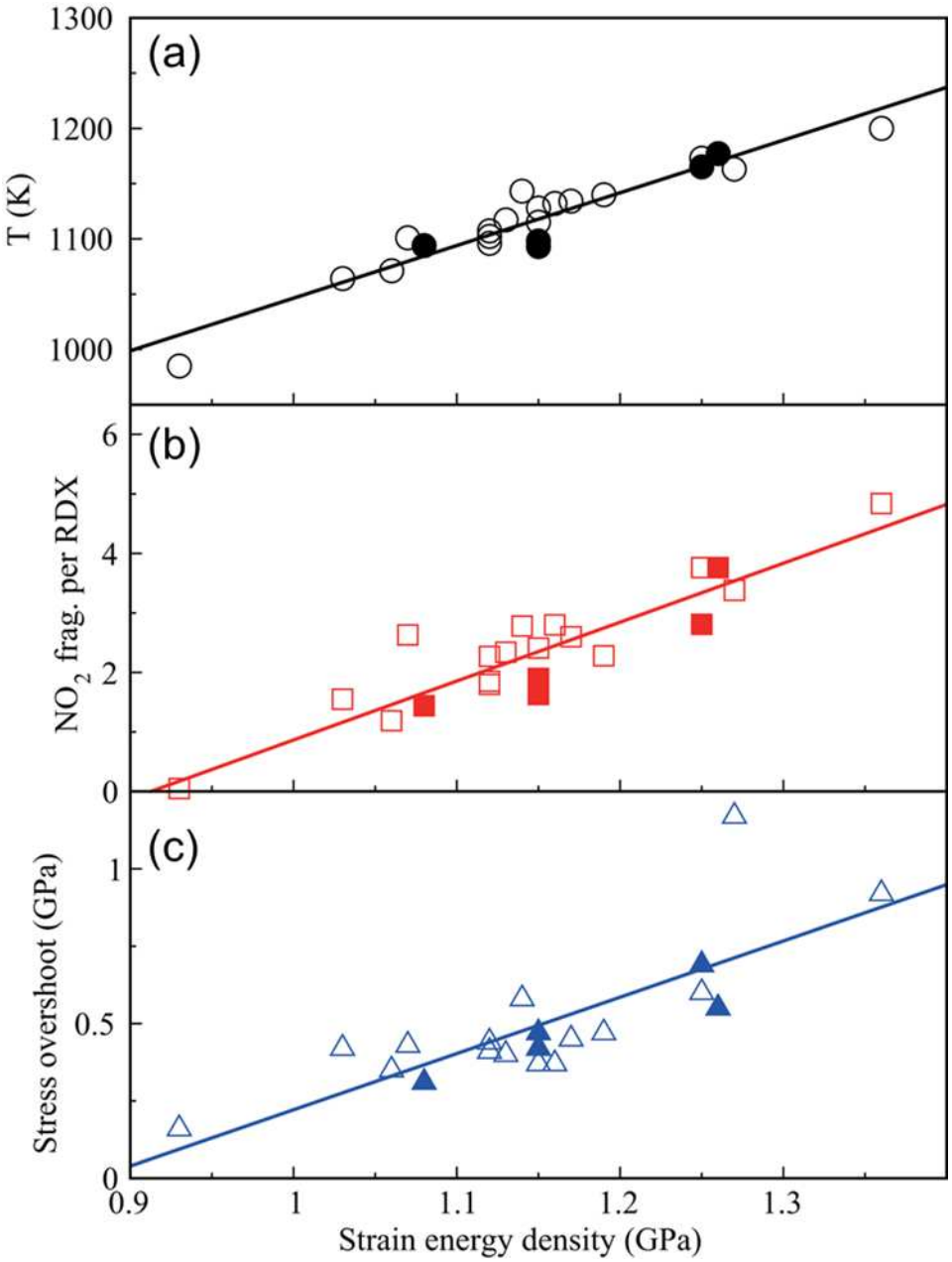

Figure S-1. Correlations between strain energy density [integration of shear stress up to 4.0 ps in the figure 2 (b)] and other sensitivity measurements for all the CS-RD cases for 10% pre-compression: (a) temperature at 8 ps, (b) NO_2 production at 10 ps, (c) stress overshoot during CS-RD. The strain energy density shows a linear relationship with other sensitivity measurements. The filled symbols are the results of selected possible slip system for the five shock planes shown in figure 3.